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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Up/Down trend in the MODIS Aerosol Optical Depth and its relationship to the Sulfur Dioxide Emission Changes in China during 2000 and 2010" *by* S. Itahashi et al.

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Thank you very much for kind reviewing of our manuscript. We have revised our manuscript according to your comments and suggestion. We believe that we have made sufficient revision to the revised manuscript. We will provide a point-by-point response below.

Reviewer Comments

This manuscript is about an analysis of the inter-annual variations of SO2 emissions from China between 2000 and 2010. Authors insisted that the SO2 emissions should increase at a rate of 12.7%/yr until 2005, and have then decreased at an average rate

of -3.9%/yr. In order to prove this, authors analyzed fine-mode AOD (AODf), REAS SO2 emissions, CMAQ modeling with the REAS SO2 emissions, and SO2 VCDs (SO2 VCDs from Gottwald and Bovensmann, 2011). This paper is dealing with an interesting and important topic in the air quality of East Asia. Also, the topic is suitable for ac&p. Therefore, it can be published on ac&p, but it also needs several modifications and improvements, before its publication on ac&p.

General comments,

1) What is the real use of the CMAQ modeling? The CMAQ modeling was performed only until 2005. Thus, it could not show (or reproduce) the decreasing trend from 2006 to 2010. Then, what is the point of using the CMAQ model in this study? This reviewer recommends to remove it from the analysis. Or, alternatively authors may be able to use only the SO2 bottom-up emission data in this analysis.

Thank you for your helpful suggestion. We used CMAQ modeling system due to the following three reasons. 1) MODIS satellite data provide the fine-mode AOD (summation of sulfate, nitrate, elemental carbon, organic carbon, and others), however, by using the CMAQ modeling system, we could know the information of aerosol component. From this point, we analyzed the contribution ratio of sulfate aerosol to fine-mode AOD (P21979, L18-20) and confirmed that the contribution ratio of aerosol sulfate to the AODf is more than 70 % on annual mean. 2) The spatial distribution and chemical process of aerosol concentration are affected by the meteorological conditions in each year, however, we evaluated that the changes of fine-mode AOD over East Asia are mainly dominated by not the meteorological conditions, but the changes of emissions based on the two sets of numerical experiments by CMAQ modeling system (P21981, L26 – P21982, L3). 3) For further study, we try to simplified inversion estimation of emissions by combining the sensitivity analysis of model and satellite measurements (P21982, L8 – L25). On these three points, CMAQ modeling analysis is needed in this study.

2) Authors primarily analyzed AODf in this study, but this reviewer think that a direct quantity to be analyzed is NOT AODf, BUT SO2 VCD. This is because the relation between AODf and SO2 emissions is much more indirect than that between SO2 VCD and SO2 emissions. For example, SO2 is oxidized first through homogeneous and heterogeneous reaction pathways to be converted into sulfate, but still AODf is not sulfate (even if sulfate is a main contributor to AODf). Therefore, this reviewer strongly recommends that authors should use the SO2 VCD (from GOME, OMI, and SCIAMACHY) as a primary variable and AODf as secondary variable in the analysis.

Thank you for this comment. SO2 is oxidized and converted to sulfate aerosols, therefore the SO2 vertical column density (VCD) data by GOME and SCIAMACHY in the down-wind region could not be retrieved properly, because the concentration of SO2 is low. From this point, we used SO2 VCD only in Central Eastern China. In this sense, we could alternatively use the fine-mode AOD (major contributor is sulfate aerosol) data observed by MODIS. This point is added as follows:

[P21977, L29] SO2 VCD data is considered the direct related to the SO2 emissions, however, this data are only used above CEC. SO2 is oxidized and first through homogeneous and heterogeneous reaction pathways to be converted to sulfate aerosols, therefore the SO2 VCD in the down-wind region is small and could not be retrieved properly.

3) The choice of the four study regions in Fig. 1 is somewhat arbitrary. Why did not authors choose the Yellow Sea, even if it is the best region to monitor these increasing/decreasing trends of the SO2 emissions from China? Why is the number 1 box (region) so small? Why did not authors work over the whole East China Sea? This reviewer thinks that the Sea of Japan (box3) would be influenced more by the Korean emissions. Therefore, this reviewer recommends that authors analyze the changing trends of the SO2 VCD and AODf over the four large boxes, for instance, the Yellow Sea, East China Sea, Sea of Japan, and remote Pacific Ocean.

Thank you for your constructive comments. This point is also pointed out in specific comment of Referee #1 - 6. Please see the answer for this comment.

4) This reviewer is not sure that authors are aware of the GAINS-ASIA projection (by IIASA). According to the GAINS-ASIA projection, the SO2 emissions from China are expected to decrease approximately from 2015 (although it has a little plateau after 2005). Authors should make some comments on this, too.

Thank you for comment from another point of view. We checked the GAINS-ASIA projection. As commented, SO2 emission from China has decreasing trend from 2015 on the Baseline08 scenario. Though, the variation of SO2 emissions is depends on the scenario. For instance, UNEP_IEA09_REF scenario shows the SO2 turning point on 2005. These point are added in our manuscript as follows:

[P21976, L4] From a different point of view, the Greenhouse Gas and Air Pollution Interactions and Synergies (GAINS) model developed by International Institute for Applied Systems Analysis (IIASA) (http://www.iiasa.ac.at/) provide the SO2 emissions from China on the long-term projection (1990-2030), however, its perspective is depends on the scenario. To capture the contemporary status of emissions, near real-time analysis of the emissions trend is the challenging work.

Specific comments (listed as page(line))

1. Page 21973, line14-16: To use AODf in this study, authors should show a more detailed analysis about the relations between sulfate and AODf possibly, from their CMAQ modeling results).

We added the some explanation about this point as follows:

[P21974, L15] we confirmed that the contribution ratio of aerosol sulfate to the AOD is more than 70 % on annual mean and . . .

2. Page 21976, line 2-4: Again, the SO2 emissions are more directly related with SO2 VCD than AODf.

On the response of General Comments 2), we used AODf on our analysis instead of SO2 VCD in the down-wind region, due to the SO2 VCD has very small value on the down-wind region.

3. Page 21978, line 17-1: In this reviewer's best knowledge, the CMAQ model does not contain the stratospheric chemistry. The contributions of stratospheric particulate species to AODf would be small. Authors should re-write this sentence.

Thank you for your comment. We added some descriptions for the aerosols of stratosphere as follows:

[P21979, L3] AOD is calculated from the all vertical layers of our modeling system. From the analysis of vertical distribution, we confirmed that the aerosol concentration are mostly high in the lower troposphere, especially within the planetary boundary layer.

Also, we excluded the text of P21981, L5-L6. Because the Vernier et al. (2011) reported that the this fluctuations is would be caused by the volcanic activity.

4. Page 21979, line 13-18: AODf from MODIS and AODf from CMAQ should be compared each other more clearly, even though it was shown briefly in Fig. 2(a) and (b). This point is also noted by the Referee #1, and please see the answer for specific comment of #1-6.

5. Page 21981, line17: Again, if authors analyze the SO2 VCDs, then authors can also analyze the increasing/decreasing trends over CEC (over the land. This reviewer believe that it can produce more direct and concrete evidences on the authors' points in this manuscript.

At first, we apologize that insufficient explanation made you confused. We used satellite retrieved SO2 vertical column density (VCD) data by GOME and SCIAMACHY to evaluate the temporal variations of SO2 emissions above Central Eastern China. From this analysis, we confirmed that the temporal variations of SO2 emissions and SO2

VCD in China are well correspond to the fluctuations of fine-mode AOD above the down-wind region (Sea of Japan). These analyses are summarized in Figure 4.

6. Page 21989, Fig. 1: Color scheme should be improved. A pink-like color for >80 Kt/yr grid is not very suitable for such high SO2 emissions.

We modified the color scale for better visualization.

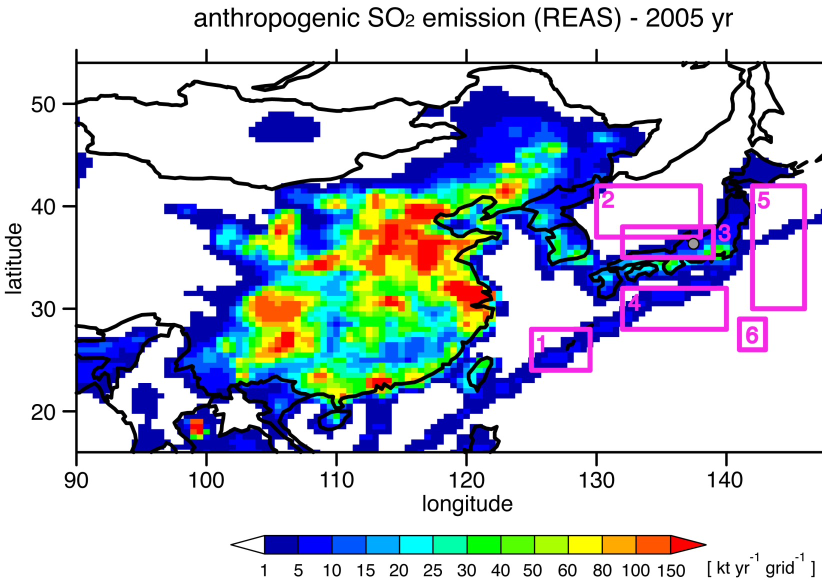
7. Page 21990, Fig. 2: Characters and scale bars inside the panels are too small to read. Again, why were 3-year averages calculated in Fig. 2(a), but 6-year averages in Fig. 2(b)? How did authors calculate the changes in AODf in Fig. 2(c) and (d)? Subtraction or division? Authors have to explain.

Thank you for your careful review of our manuscript. This point is also noted by the Referee #1, and please see the answer for specific comment of #1-6.

8. Page 21991-21992, Fig. 3 & Fig. 4: Again, characters in Fig. 4 are too small to read. Two figures should be replaced or re-plotted, with the major points 2 & 3 above.

Thank you. We revised of this point for better reading.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 21971, 2011.



1. South of East China Sea
2. Sea of Japan
3. Mt. Tateyama
4. South of Japan
5. East of Japan
6. Ogasawara

● ground observation site;
Mt. Tateyama in Murododaira
(36.57°N, 137.60°E, 2450 m)

Fig. 1. revised figure of Fig. 1